

### **Remarks**

Claims 1, 17, and 19 are amended. Claims 1 to 24 are pending.

### **112 Rejections**

Claims 4, 5, 7, 8, 10, 11, 20, 21, and 22 were rejected under 35 USC § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which Applicants regard as the invention.

The Patent Office submits that the claims are indefinite because it is unclear what is meant by "epoxy equivalent weight" and "hydroxy equivalent weight" as used in the claims.

Applicants respectfully disagree with the Patent Office that the claims are indefinite. Epoxy equivalent weight and hydroxy equivalent weight are terms that are known in the art of epoxy resins and are used to characterize epoxy resins. Epoxy equivalent weight is defined as the weight of resin in grams which contains one gram equivalent of epoxy. Hydroxy equivalent weight is the weight of resin containing one equivalent weight of hydroxyl group. Epoxy equivalent weight and hydroxy equivalent weight can be determined by several methods such as by wet analysis and near-infrared spectroscopy. Definitions of these terms and a discussion of the methods by which they can be determined are available in technical publications such as Lee and Neville's Handbook of Epoxy Resins, Chapter 4, Characterization of Uncured Epoxy Resins, pages 4-14 and 4-21 (McGraw-Hill, Inc., 1967). A copy of the cited pages accompanies this response.

Applicants respectfully request reconsideration and withdrawal of the above rejection of claims 4, 5, 7, 8, 10, 11, 20, 21, and 22 in view of the discussion above.

### **103 Rejections**

Claims 1-5, 12-20, and 23-24 were rejected under 35 USC § 103(a) as being unpatentable over Welke et al. (EP 1026218 A1) in view of Pike (US 6,352,948). The Patent Office submits in part that: Welke et al. discloses a UV-curable polyester/epoxy adhesive tape that is capable of being cured upon exposure to actinic radiation; adhesive tapes that comprise at least one backing which can include a non-woven material; and that since the adhesive is coated on the non-woven in a molten state, it would be probable that the adhesive would saturate the material, partially

embedding the backing. The Patent Office also submits that Pike teaches a fine fiber composite web laminate having first and second fibers having specified cross sectional areas.

Applicants have discovered a way to control cold flow of the uncured adhesive by incorporation of a web in the adhesive. Applicants show by way of example that inclusion of the web decreases the cold flow of uncured adhesive tape when subjected to a load of two pounds for 72 hours at room temperature (page 65, Table 7).

Applicants now claim s structural adhesive layer comprising an adhesive matrix and a web that is completely embedded within the outer surfaces of the adhesive layer. Applicants have discovered that webs having a particular air permeability and light permeability as specified in the present claims when embedded within an adhesive layer, improve (reduce) uncured adhesive cold flow. Additionally, such webs do not negatively impact the ability of curing radiation to reach portions of the structural adhesive layer on an opposite side of the fiber reinforcement.

Welke et al. disclose pressure sensitive adhesive tape containing a UV-curable epoxy/polyester resin mixture. The tapes may be unsupported transfer tapes or supported tapes (paragraphs 62-63). Supported tapes have at least one backing. The adhesive may be applied to the backing by coating a molten mixture of adhesive onto a backing or laminating an adhesive transfer tape to a backing. Welke et al. do not suggest embedding a web within an adhesive layer.

Pike discloses fine fiber nonwoven composite web attached to a barrier layer thermally, ultrasonically, mechanically, and/or adhesively. Pike discloses that the layers may be bonded together by applying an adhesive between the layers (column 9, lines 25-26). Pike is silent with regard to any air permeability requirement nor suggests imbedding such nonwoven composite within an adhesive layer.

Even assuming that the disclosures of Welke et al. and Pike are combinable as suggested by the Patent Office, such a combination as suggested would not result in the present invention as now claimed. At most, such a combination would result in a structure having an epoxy/polyester

adhesive bonded to a nonwoven composite web. Clearly, this is not the invention as presently claimed. For at least this reason, and assuming such combination is proper, the combination of Welke et al. and Pike as suggested by the Patent Office does not result in the claimed invention and thus, cannot render the claimed invention obvious. Accordingly, Applicants respectfully request that the above rejection of claims 1-5, 12-20, and 23-24 be withdrawn.

Claims 1, 6-8, 19, and 21 are rejected under 35 USC 103(a) as being unpatentable over Willett (WO 99/57197) in view of Pike (US 6,352,948) and Pachl et al (US 6,174,932). The Patent Office submits in part that: Willett discloses a curable composition formed by mixing components comprising curable epoxy resin, curative for the epoxy resin, thermoplastic ethylene-vinyl acetate copolymer and thermoplastic polyester resin and that the curable composition may be applied to a variety of substrates including plastics, metals, ceramics, glass and cellulosic materials; Pike teaches a fine fiber composite web laminate having first and second fibers having specified cross sectional areas; and Pachl et al. disclose a UV curable sealant composition having at least one epoxy, at least one polyol, at least thickener, at least one monomer, and at least one phenoxy resin.

Willett '197 discloses curable compositions containing epoxy resin, ethylene-vinyl acetate copolymer, and polyester resin that may be supplied as an unsupported film or may be applied to substrates such as cellulosic materials and metals. Pike has been discussed above. Pachl et al. disclose curable sealant compositions that may be applied to a substrate such as wood, steel, glass, or cement.

None of Willett '197, Pike, or Pachl et al. teach or suggest an embedded web within outer surfaces of an adhesive layer. Thus, the combination of such disclosures (assuming such combination is proper) does not teach or suggest the invention as claimed. For at least this reason, such combination as suggested by the Patent Office cannot render the instantly claimed invention obvious. Accordingly, Applicants respectfully request that the above rejection of claims 1, 6-8, 19, and 21 be withdrawn.

Claims 1, 9-11, 19, and 22 are rejected under 35 USC 103(a) as being unpatentable over Nakasuga et al. (US 6,376,070) in view of Willett et al. (US 6,136,398) and Pike et al. (US 6,352,948). The Patent Office submits in part that: Nakasuga et al. is directed to sheet-form, curable pressure sensitive adhesive sheet and discloses an adhesive comprising a high molecular weight polymer, a compound containing an epoxy group and a polymerization initiator; the sheet-form, curable pressure-sensitive adhesive may be placed on at least one surface of a substrate such as rayon or cellulosic non-woven fabrics or sheets made of synthetic resins such as polyethylene, polyester, polypropylene and polystyrene; Willett teaches a curable adhesive composition comprising epoxy resin, curative for the epoxy resin, ethylene-vinyl acetate copolymer, thermoplastic polyester resin, various additives, and may comprise a hydroxyl-containing material; and Pike teaches a fine fiber composite web laminate having first and second fibers having specified cross sectional areas.

Willet et al. '398 is the priority application of Willett '197, and thus, has been discussed above. Pike has been discussed above. Nakasuga et al. disclose a sheet-form pressure sensitive adhesive containing an acrylic polymer, a compound having an epoxy group, and a cationic polymerization initiator. Nakasuga et al. disclose that such a sheet-form adhesive may be placed on at least one surface of a substrate to form a curable pressure sensitive adhesive sheet.

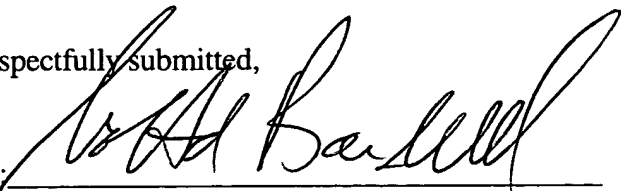
None of the above references disclose or suggest an embedded web within outer surface of an adhesive layer. Thus, assuming such references are properly combinable as suggested by the Patent Office, such combination would not result in the invention as presently claimed or provide a tape having improved cold flow characteristics. For at least this reason, the suggested combination of references, if assumed to be proper, cannot render the claimed invention obvious. Accordingly, Applicants respectfully request that the above rejection of claims 1, 9-11, 19, and 22 be withdrawn.

In view of the above, it is submitted that the application is in condition for allowance.  
Reconsideration of the application is requested.

9 September, 2003  
Date

Respectfully submitted,

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## OTHER MCGRAW-HILL HANDBOOKS OF INTEREST

BAUMEISTER AND MARKS · Standard Handbook for Mechanical Engineers  
 BEEMAN · Industrial Power Systems Handbook  
 BELL · Petroleum Transportation Handbook  
 BLAND AND DAVIDSON · Petroleum Processing Handbook  
 BLATZ · Radiation Hygiene Handbook  
 CARROLL · Industrial Instrument Servicing Handbook  
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 GUTHRIE · Petroleum Products Handbook  
 HEYEL · The Foreman's Handbook  
 HUSKEY AND KORN · Computer Handbook  
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 KATZ · Handbook of Natural Gas Engineering  
 KING AND BRATER · Handbook of Hydraulics  
 KLERER AND KORN · Digital Computer User's Handbook  
 KORN AND KORN · Mathematical Handbook for Scientists and Engineers  
 LANGE · Handbook of Chemistry  
 LASSER · Business Management Handbook  
 MAGILL, HOLDEN, AND ACKLEY · Air Pollution Handbook  
 MANAS · National Plumbing Code Handbook  
 MANTELL · Engineering Materials Handbook  
 MAYNARD · Industrial Engineering Handbook  
 MAYNARD · Top Management Handbook  
 MEITES · Handbook of Analytical Chemistry  
 MOODY · Petroleum Exploration Handbook  
 MORROW · Maintenance Engineering Handbook  
 PERRY · Chemical Engineers' Handbook  
 PERRY · Engineering Manual  
 RICHEY, HALL, AND JACOBSEN · Agricultural Engineers Handbook  
 SHAND · Glass Engineering Handbook  
 STANIAR · Plant Engineering Handbook  
 STREETER · Handbook of Fluid Dynamics  
 TOULOUKIAN · Retrieval Guide to Thermophysical Properties Research Literature  
 TRUXAL · Control Engineers' Handbook

# HANDBOOK OF EPOXY RESINS

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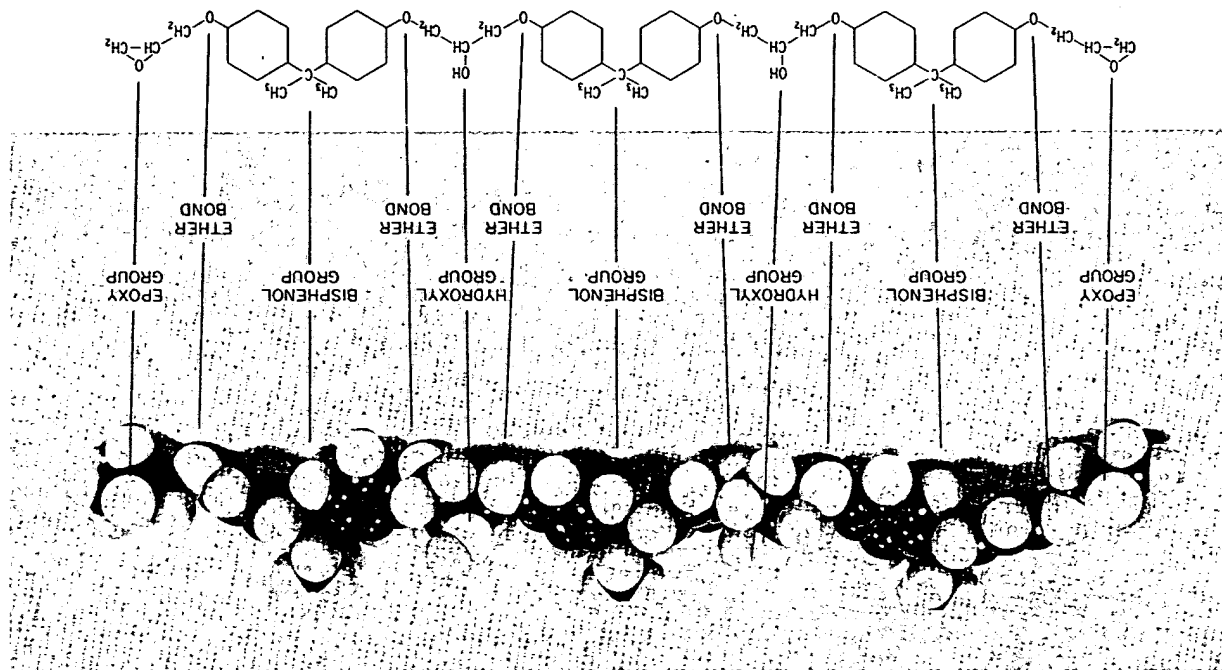
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# HANDBOOK OF EPOXY RESINS

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Atom model of widely used solid epoxy resin, diglycidyl ether of bisphenol A, degree of polymerization of 2 (DGEBA,  $n = 2$ ). Approximate magnification: 100 million times. (*The Epoxylite Corporation*.)

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## VISCOSITY

Viscosities of liquid resins are usually measured with a rotating spindle instrument, such as the Brookfield viscometer. A typical procedure is indicated in Table 4-9. Viscosity vs. temperature for various of the liquid epoxy resins is indicated in Fig. 4-8. The sharp dependence of viscosity on temperature should be noted.

Solid resins are usually dissolved to a 40 per cent solution in butyl Carbitol. Solution viscosity is measured by spindle viscometers, by bubble viscometers such as the Gardner-Holdt tubes, or by the capillary viscometers, such as the Ostwald. Viscosity data for various solid resins in solution are given in Chap. 24.

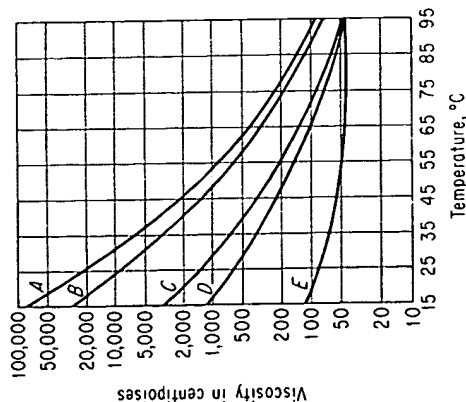
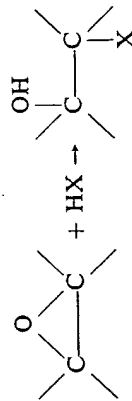


FIG. 4-8. Viscosity vs. temperature of five types of epoxy resins. *A*, Epoxidized phenol-formaldehyde novolac (*f* 2.3); *B*, DGEBA; *C*, 3,4-epoxy-6-methylcyclohexylmethyl-3,4-epoxy-6-methylcyclohexane carboxylate; *D*, diglycidyl ester of linoleic acid; *E*, DGE polypropylene glycol.

## Determination of Epoxide Equivalent by Wet Analysis

The most frequently reported and, in general, the most specific and practical methods for determination of epoxide equivalents are based on the addition of a hydrogen halide to the epoxy group.



The difference between the amount of acid added and the amount unconsumed, determined by titration with standard base, is a measure of the *epoxy content*.

In practice, hydrogen chloride, hydrogen bromide, or hydrogen iodide is used (Table 4-10). One procedure involves the use of hydrogen bromide generated *in situ* from perchloric acid and a quaternary ammonium bromide and is reported to provide faster titrations and sharper end points than with HBr-acetic acid [45]. The four most common methods are the pyridinium chloride-pyridine method, the hydrogen bromide-acetic acid method, the hydrochloric acid-potassium iodide

Table 4-9. Test Method for Viscosity of Epoxy Compounds [32]

### Apparatus:

Brookfield viscometer, model RVF  
Thermometer, centigrade with 0.1° divisions  
600-ml glass beaker or quart open-top paint can

### Procedure:

1. Place 500 ml of the sample to be tested in the clean beaker or can. Insert the selected spindle and guard into the sample, being careful to avoid the trapping of air under the spindle plate.
2. Bring the sample to  $25 \pm 0.1^\circ\text{C}$ .

### Testing:

1. Attach the spindle and guard to the Brookfield viscometer, and adjust the spindle depth to the notch on the spindle shank.
2. Start the viscometer. Allow the spindle to rotate for  $\frac{1}{4}$  min. Stop the indicator by use of the clutch. Then stop the instrument, and read the dial. Allow the spindle to rotate an additional 3 to 4 times, and take a second reading. If the original reading agrees with the second reading, record this figure. If, however, the readings differ, continue spindle rotation for a reasonable period of time until a constant value is obtained.
3. Convert the reading obtained from the dial to viscosity in centipoises in accordance with the conversion table which accompanies the Brookfield viscometer.
4. Record the following information for each determination:
  - a. Viscosity in centipoises
  - b. Spindle
  - c. Speed
  - d. Temperature of sample to the nearest  $0.1^\circ\text{C}$
  - e. Ambient room temperature to the nearest  $0.5^\circ\text{C}$

method, and potentiometric titration with hydrochloric acid. Procedures for conducting these methods are detailed in Tables 4-11 to 4-14.

The applicability of each method depends somewhat on the resin type being analyzed, as well as on other factors. First, the reagents should provide good solvent properties for the sample, particularly if the sample is a solid. Thus, the aqueous magnesium chloride-hydrochlorination method has a limited scope because only such epoxies as glycidol are readily soluble in the aqueous salt solution although less soluble epoxies may dissolve slowly and consequently may react at somewhat slower rate. The alcoholic magnesium chloride-hydrochlorination method is more general, in that many higher epoxies, such as liquid aromatic glycidyl ethers, are soluble in the reagent. However, high-molecular-weight epoxies such as the more highly polymerized aromatic glycidyl ether epoxy resin are not soluble in the alcohol medium. The hydrochloric acid-ethyl ether reagent is a good solvent for many monoepoxies but is not satisfactory for epoxy resin. The hydrochloric acid-cellosolve reagent is a fairly good solvent for epoxy resin; but most samples go into solution rather slowly, requiring heating and mixing over a considerable length of time. Pyridine, chloroform, and dioxane are all good solvents for epoxies, including aromatic glycidyl ether resins.

Another factor in the applicability of a method depends on the extent to which undesirable side reactions occur. Any reaction of the epoxy is undesirable which does not result in the net consumption of one mole of acid per equivalent of epoxy group. For example, under certain conditions, some epoxies tend to isomerize to the corresponding carbonyl compounds and some of the epoxies or their chlorohydrins are susceptible to hydrolysis or alcoholysis. These reactions offer competition for the desired hydrochlorination reaction.

Various other organic radicals present in commercial resins may interfere with the desired reaction. For example, phenols interfere in the pyridinium chloride chloroform method by causing poor end points; organic acids lower precision accuracy, and in some methods various esters partially hydrolyze. A detailed study of such interfering factors is found in [12].



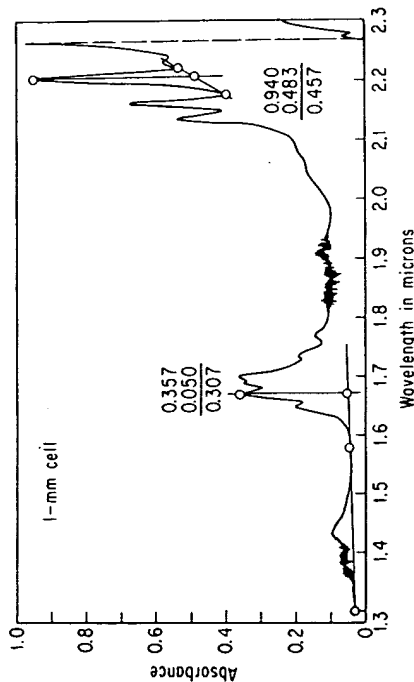


FIG. 4-10. Near-infrared spectrum of DGEBA showing evaluation of epoxy peak at 2.05 microns and reference peak at 1.668 microns [34].

Table 4-16. Near-Infrared Absorbance for Epoxy Groups [34]

DGEBA	Density, $d_{20}^4$	Epoxy value by wet analysis			Absorbance		
		equiv./100 g	equiv./liter		2,205 $\mu$ , 1-mm cell	1,668 $\mu$ , 1-mm cell	1,159 $\mu$ , 10-mm cell
Crystallized	1.162	0.585	6.79		0.707	0.311	0.164
Distilled	1.165	0.573	6.68		0.685	0.311	0.161
Mol. wt. 380	1.165	0.522	6.08		0.620	0.309	0.140
Mol. wt. 450	1.163	0.395	4.60		0.457	0.307	0.090
Mol. wt. 950-1,000	1.188	0.197	2.34		0.208	0.293	0.015
Mol. wt. 1,400	1.177	0.099	1.164		0.221	0.937	0.184

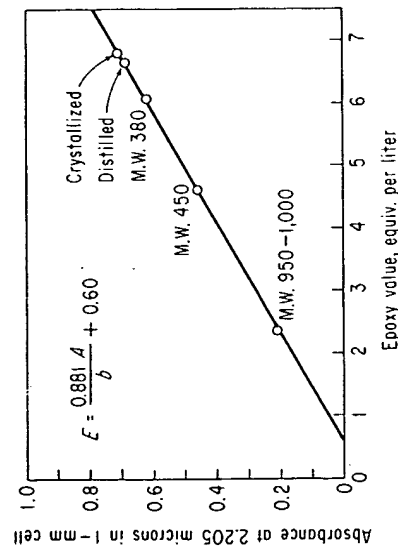


FIG. 4-11. Determination of epoxy values (direct method) vs. molecular weight of DGEBA [34].

A well-defined peak for  $\alpha$ -epoxies is that at 2.205 microns, which probably represents a combination of a C—H stretching fundamental ( $\nu = 3,050 \text{ cm}^{-1}$ ) with a CH deformation ( $\nu = 1,460 \text{ cm}^{-1}$ ) [34]. A typical near-infrared absorption spectrum for the DGEBA is shown in Fig. 4-10. A calibration curve, comparing

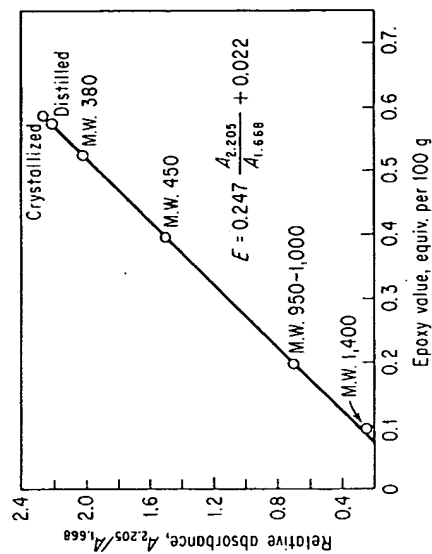


FIG. 4-12. Relative absorbance in near infrared vs. molecular weight of DGEBA [34].

epoxy value determined by wet analysis with absorbance at 2.205 microns, for five epichlorohydrin-bisphenol A resins is presented in Fig. 4-11. The data are tabulated in Table 4-16.

Indirect methods have also been used for determining epoxide equivalent. The variable absorbance of the epoxy group can be compared against a relatively nonvariable absorbance, such as the first overtone of the aromatic carbon-hydrogen stretching mode at 1.668 microns for the DGEBA resins [34]. Data for such a correlation are given in Table 4-16 and Fig. 4-12.

The absorption band at 1.159 microns has also been employed to determine epoxide equivalent (Fig. 4-13).

## HYDROXYL EQUIVALENT

Hydroxyl equivalent is the weight of the resin containing one equivalent weight of hydroxyl group. It may also be expressed as equivalents per 100 grams. Hydroxyl equivalents may be determined by several methods, the most common being (1) esterification with acids, (2) reaction with acetyl chloride, (3) reaction with lithium aluminum hydride, and (4) reaction with phenyl isocyanate. Near-infrared spectroscopy may also be used.

Wet analysis, such as the benzyltrimethylammonium periodate method, is used for determining  $\alpha$ -glycol content of the epoxy resins.

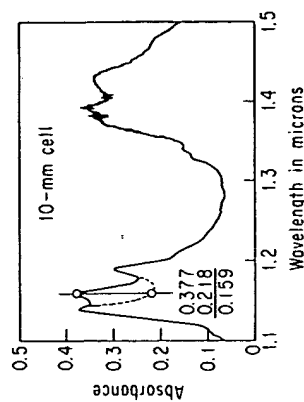


FIG. 4-13. Infrared spectrum of DGEBA showing evaluation of epoxy peak at 1.159 microns [34].